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Abstract

# FLOURIDE VOLATILITY PROCESSES FOR THE RECOVERY OF FISSIONABLE MATERIAL FROM IRRADIATED REACTOR FUELS

Richard C. Vogel (Argonne National Laboratory)
W. H. Carr (Oak Ridge National Laboratory)
G. I. Cathers (Oak Ridge National Laboratory)
J. Fischer (Argonne National Laboratory)
L. P. Hatch (Brookhaven National Laboratory)
R. W. Horton (Oak Ridge National Laboratory)
A. A. Jonke (Argonne National Laboratory)
R. P. Milford (Oak Ridge National Laboratory)
J. J. Reilly (Brookhaven National Laboratory)
G. Strickland (Brookhaven National Laboratory)

In flouride volatility processes the fissile material in spent nuclear fuels is converted into volatile hexafluorides that can be separated from fission products and other materials by volatilization. Possible advantages are (1) fewer processing steps, (2) the use of reagents with low susceptibilities to deleterious radiation effects, (3) a uranium product (uranium hexafluoride) in the form desired for isotope separation, (4) radioactive waste products in solid form and small volume, and (5) a small shielded-space requirement. These processes are being developed for both high-and low-enrichment fuels.

In fused-salt processes for enriched fuels, the fuel is dissolved at 650C or less in a molten fluoride mixture by bubbling hydrogen fluoride through the melt, The uranium is then fluorinated to the hexafluoride at 500C and volatilized. The hexafluoride is decontaminated from fission products by a sorption-desorption cycle using sodium fluoride as the asborbant.

In a recent series of hot pilot-plant runs, highly decontaminated uranium hexafluoride was recovered. The highest individual decontamination factor (around $10^{10}$) was for zirconium and the lowest was for ruthenium (around $10^{8}$).

In the fluidized-bed fluoride volatility processes, the spent fuel material is submerged in a bed of fluidized inert alumina while dry gases are reacted with the fuel. The fluidized bed dissipates the heat of reaction. This process, when applied to enriched zirconium or aluminum fuels, has the following major steps:

(1) Hydrogen chloride is reacted with either zirconium (at about 400C) or aluminum. The zirconium or aluminum chlorides are converted to the oxides in a separate fluid bed by reacting with steam. (2) The uranium and fission-product chlorides remaining in the alumina bed are converted to the fluorides by hydrogen fluoride. (3) These fluorides are then fluorinated and the volatilized uranium hexafluoride is collected in cold traps. (4) Further decontamination can be achieved by distillation or by the procedures described above. This process is now being demonstrated in cold pilot-plant experiments.

Work is being done on the fluidized-bed fluoride volatility process for low enrichment fuels of uranium dioxide clad in Zircaloy or stainless steel. These fuels contain plutonium that must be recovered. The present process plan has the following major steps: (1) The uranium and the plutonium are separated from the fuel jacket. For zirconium, this separation has been done on an engineering scale with hydrogen chloride by formation and volatilization of the zirconium tetrachloride. A possible procedure for stainless steel cladding consists of removing the oxide from the chopped cladding through formation of powdered $U_3O_8$ by oxidation of the dioxide. (2) The oxides are fluorinated with fluorine gas at about 500C. (3) Distillation of the hexafluorides may be used to remove fission products and to partition the hexafluorides into uranium hexafluoride and a mixture more concentrated in plutonium. (4) The mixture of plutonium and uranium hexafluorides can be converted by reacting with hydrogen and steam to the oxides, which can then be fabricated back into reactor fuel.

The fluid-bed process has been partially demonstrated on an engineering scale using non-radioactive materials. Bench-scale studies show a potential for application to plutonium-bearing fuels. Facilities for pilot-scale demonstration using plutonium and tracer levels of fission products are nearing completion.

Additional versatility of the fluid-bed method for carrying out gas/solid reactions has been demonstrated by recovering uranium hexafluoride from graphite matrix fuels. At the present time a pilot plant is under construction to demonstrate this process for graphite-matrix fuels.

The technology of fluoride volatility processes is nearing the stage at which design can be started for a commercial-scale plant for enriched-uranium fuels. Additional development work is necessary to confirm the promising prospect for application of these schemes to low-enrichment power-reactor fuels containing plutonium.